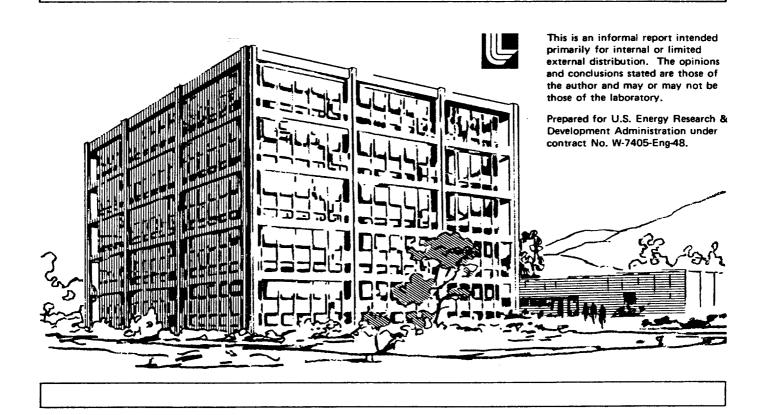
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THE SPREADING AND DIFFERENTIAL BOIL-OFF FOR A SPILL OF LIQUID NATURAL GAS ON A WATER SURFACE

WERNER STEIN

AUGUST 1978

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THE SPREADING AND DIFFERENTIAL BOIL-OFF FOR A SPILL* OF LIQUID NATURAL GAS ON A WATER SURFACE

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ABSTRACT

A model for the spreading and evaporation of liquid natural gas (LNG) when spilled on a liquid surface has been developed. The model includes a model for differential boil-off of the LNG constituents. A listing of the code, LNGVG, for making these calculations plus calculational results for an anticipated LNG spill test to be conducted at China Lake, California are included in the documentation.

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INTRODUCTION

Liquid natural gas (LNG) is a cryogen with a boiling temperature, at l atm., of approximately 111° K. Spillage of LNG on a water surface results in a very rapid spreading into a circular shaped pool. During this spreading process, heat transfer from the relatively warm water to the cold LNG results in boiling of the LNG with a resultant high rate of gas vapor generation.

LNG is composed primarily of methane with small fractions of ethane, propane and nitrogen, see page 5. These constituents have different heats of vaporization and boiling points with the result that they boil off at different rates. This differential boil off during vapor generation results in LNG vapors containing different fractions of constituents than the originally spilled LNG.

A computer program called LNGVG to calculate <u>LNG Vapor Generation</u> and differential boil off has been written. A listing of the code is given in Appendix A. Calculations have been made, using LNGVG, for a spill of 5 cubic meters of LNG in a time of 50 seconds and are described below. These spill conditions are representative of conditions expected during experiments to be conducted at China Lake.

DISCUSSION

The calculations for the spreading of LNG are approached by determining the velocity of the leading edge of the LNG pool. This velocity is determined by considering the outer edge as a density intrusion. The radius of the pool as a function of time is determined by integration of the velocity equation. Spreading occurs until pool break-up occurs.

Pool break-up is assumed to occur when the thickness of the LNG reaches an experimentally determined minimum thickness, due to spreading and evaporation. The break-up is observed to occur at the center of the pool first and spreads radially outward until all the LNG has evaporated.

The rate of evaporation for LNG from a water surface is often determined experimentally and given as a regression rate (i.e., cm./min.). This regression rate represents the sum of the contributions from each of the LNG constituents. The fraction of this regression rate applicable to each of the constituents is determined in the calculation by the relative magnitudes of their heats of vaporization, boiling temperature and volume fraction. This relative fraction of total boil-off for each constituent varies with time and is different from the original volume fraction of the LNG.

LNG SPILL SCENARIO

The phenomena that occur during a spill are described below. The LNG is assumed to be spilled at a certain rate for a finite time.

- (1) Initially the LNG spreads radially at a rapid rate, which decreases as the radius increases. Boil-off of the LNG takes place as soon as the LNG contacts the water surface. Due to the differential boil-off phenomenon, the vapors generated have different volume fractions than the initial LNG. Also, the volume fraction of the LNG constituents of the LNG on the water surface changes due to the differential boil-off.
- (2) The LNG spreads out to a radius large enough to vaporize an amount of LNG equal to the rate of LNG spill. The LNG composition on the water surface continues to change due to differential boil-off until a condition is reached where the rate of boil-off of each LNG constituent equals the rate of spillage of each constituent.
- (3) Steady conditions are maintained until the spillage is stopped. Now, the volume fraction of each constituent in the spilled LNG changes due to differential boil-off and the vapors generated again have

Volume fractions different from the original LNG volume fractions.

(4) The volume of LNG left on the water decreases as vapor generation takes place until the pool begins to break up in the center. Initially just a small circular area of water is visible. This circular area increases with time until the entire mass of LNG has evaporated.

ANALYTICAL RELATIONS

The radius of the LNG spilled on the water surface is given by equation (1):

(1)
$$r = 1.35 \left(g \frac{\rho_W - \rho_{LNG}}{\rho_W}\right)^{1/4} V t^{1/2}$$

where: r = radius

 ρ = density of LNG or water (w)

V = volume of LNG on water surface

t = time

The velocity of the leading edge of the LNG is given by differentiating equation (2) with respect to time while holding V constant:

(2)
$$\left(\frac{dr}{dt}\right)_{V=constant} = \frac{1.35}{2} \left[g \frac{\rho_W - \rho_{LNG}}{\rho_W} \right]^{1/4} v^{1/4} t^{-1/2}$$

The method of applying the above equations was to calculate the spreading for very short time increments using a constant LNG volume during the time increment. After each time increment, the volume remaining was adjusted to account for LNG added during spilling and loss due to evaporation. Thus, the radius after N+1 successive time intervals, Δt , is given by:

(3)
$$r_{N+1} = r_N + \left(\frac{dr}{dt}\right)_{V=V_N} \Delta t$$

and the volume V_N is given by: [1]

(4)
$$V_N = V_{N-1} + [\hat{V} - EV_{N-1}] \Delta t$$

where: \dot{V} = rate of addition of LNG

EV = Rate of evaporation of LNG

In the case of a continuous LNG spill, the maximum radius of the pool is given by:

$$\dot{V} = \pi R^2 K$$

where: R = maximum pool radius

K = LNG regression rate (length/time)

After spillage of the LNG has stopped, the maximum radius, R, attained by the pool is assumed to remain constant. During this condition, evaporation takes place until the average LNG pool thickness, h, equals 0.183 cm. [2] and thereafter pool break-up occurs. Pool break-up initially occurs at the center of the pool and spread radially outward as LNG is evaporated during pool break-up, the LNG thickness is assumed to remain constant at 0.183 cm. This value has been experimentally obtained with other researchers [3] obtaining different values, up to a factor of 3 larger. Using larger values for pool break-up results in pool break-up occuring sooner.

Also experimentally obtained is the rate of LNG boil-off expressed as a regression rate in units of, for example, cm. of LNG per second. A value

of 0.0423 cm. per second $^{[4]}$ was used in the subsequent China Lake calculation. This rate represents the sum of the regression rates for each of the various constituents of the LNG. The regression rate for any individual constituent, I, is calculated as follows with I = 1 corresponding to methane (CH₄):

(6)
$$K = \sum_{I} \frac{(C_{P}\Delta T + HVAP)_{CH_{4}} \rho_{CH_{4}} FRI(I)A}{(C_{P}\Delta T + HVAP)_{I} \rho_{I}} = \sum_{I} K_{I} FRI(I)$$

where K = experimentally determined LNG regression rate

 C_D = specific heat

 ΔT = number of degrees that the boiling temperature of constituent I is above the LNG boiling temperature.

HVAP = heat of vaporization

 ρ_{I} = liquid density of constituent I

FRI(I) = volume fraction of constituent I in the original LNG

A = unknown regression rate to be solved for.

Solving (6) for "A" and plugging into the below equation (7) gives the regression rate, K_T , of constituent I:

(7)
$$K_{I} = \frac{(C_{p}\Delta T + HVAP)_{CH_{4}} \rho_{CH_{4}} \Lambda}{(C_{p}\Delta T + HVAP)_{I} \rho_{I}}$$

The LNG spilled initially on the water contains various volume fractions of constituents. Throughout the calculations a mass balance is calculated for each constituent in the spilled LNG. Addition of constituents to the spilled LNG is determined from the rate of spill and the known volume fractions of the LNG. Loss of constituents from the spilled LNG is by evaporation. The amount evaporated in a time step Δt of constituent I is given by $\Delta V_{\rm I}$:

$$\Delta V_{I} = K_{I} F S \Delta t$$

where: F = volume fraction of constituent I in the LNG pool

S = surface area covered by LNG pool

In the calculations, the mixture of the constituents is always assumed to be homogeneous.

CALCULATIONS

The above relations have been incorporated into a computer code called LNGVG. A listing is provided in Appendix A. Use of this code involves generating an input file called LNGIN which contains the information called for by the read statements 6 and 8. Input variables and their units are described in the comment cards at the beginning of the code. Output is all contained in an output file called LNGOUT.

Calculations for an anticipated spill at China Lake have been made. The initial conditions for this spill are given below:

Volume of LNG spilled = 5 meter³

Rate of LNG spillage = $5 \text{ meters}^3/50 \text{ sec.}$

Volume fraction of constituents

in the LNG: Methane $(CH_4) = 0.922$

Ethane = 0.0527

Propane = 0.0112

Nitrogen = 0.0139

LNG Boiling Temperature = 111.7°K (201°R)

LNG Regression Rate = 0.0423 cm/sec (1 inch/minute)

Water Density = $1,000 \text{ Kg/m}^3$

Initial LNG Density = 439 Kg/m^3

RESULTS

The results of the calculations for the volume fraction of each constituent in the vapors generated vs. time is shown in Figures 1 through 3. From these figures, one sees that the initial volume fraction of methane and nitrogen in the vapors is greater than the original fractions in the LNG, and the initial volume fractions of ethane and propane are less than the original LNG. The volume fractions in the vapor adjust themselves with time, however, until at 50 sec (spilling stops at 50 sec) the vapors have approximately the same volume fraction as the original LNG. After spilling stops, (time greater than 50 sec) the volume fraction of methane and nitrogen decreases and that of ethane and propane increases continuously until all the LNG has evaporated.

The maximum radius attained by the LNG pool is 28.5 feet at a time of 27.0 seconds. Pool break-up is calculated to occur after 61.5 seconds and total evaporation is completed after 78 seconds.

The total boil-off rate (ft^3/sec) vs. time for this spill is shown on Figure 4.

CODE VERIFICATION

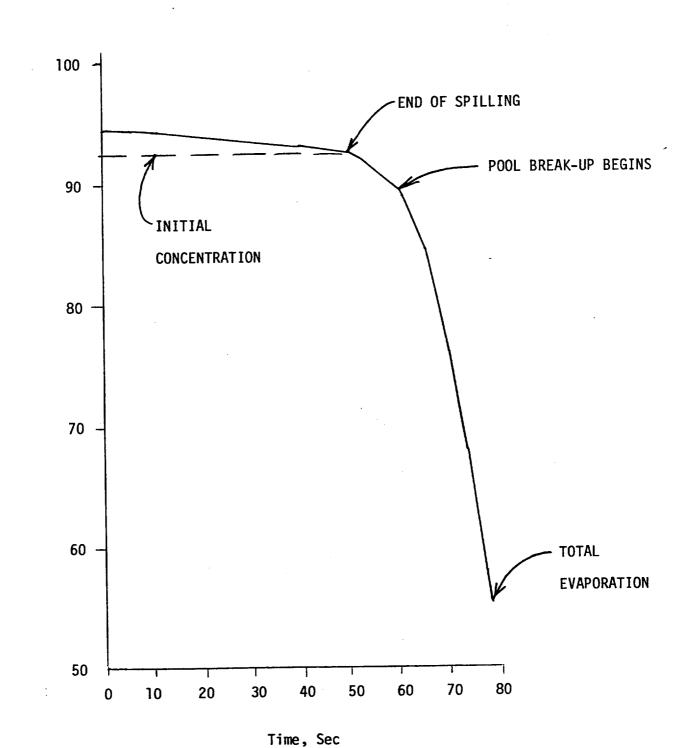
The work done in this study is primarily analytical in nature and has not been compared with experiment.

Comparisons with results of other models [4,5] have been made and are tabulated in Table I. The comparison involves maximum time to evaporate and maximum pool radius for instantaneous spills of 10 m³ and 1000 m³. The calculational results from all three models agree fairly closely.

FIGURE 1

VOLUME FRACTION OF METHANE IN LNG VAPORS

LNG SPILL SIZE = $5M^3$ SPILL RATE = $5M^3/50$ SEC



METHANE VOLUME FRACTION, %

VOLUME FRACTION OF ETHANE AND PROPANE IN LNG VAPORS

LNG SPILL SIZE = 5 M^3 SPILL RATE = $5 \text{ M}^3/50 \text{ SEC}$

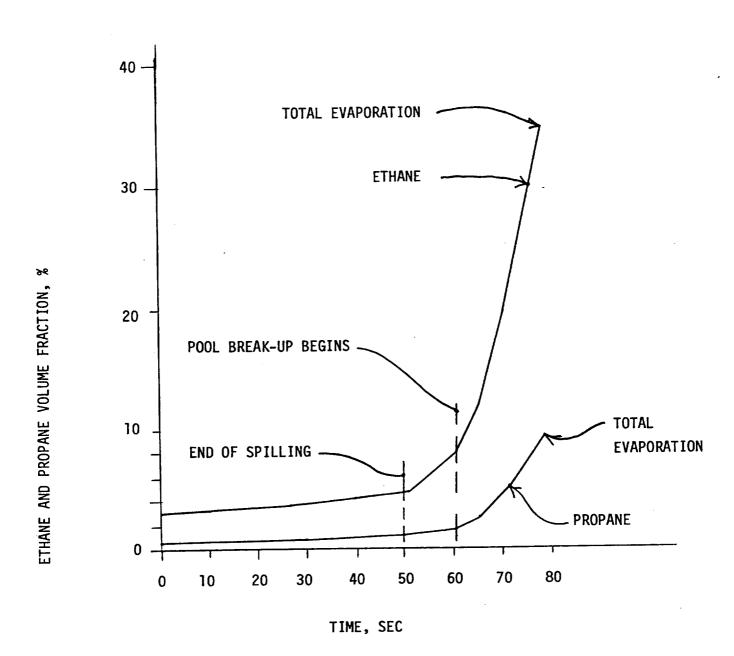
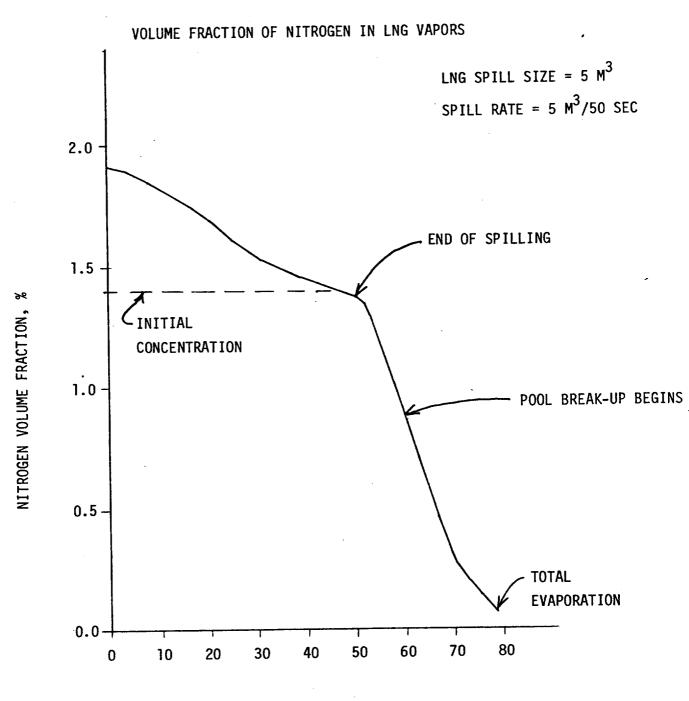


FIGURE 3

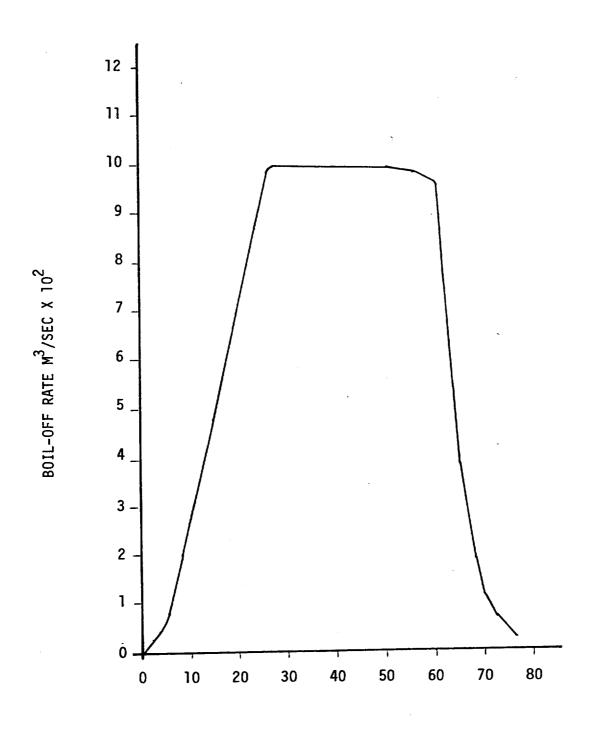


TIME, SEC

FIGURE 4

BOIL-OFF RATE FROM A SPILL OF 5 m³ OF

LNG IN A TIME OF 50 SECONDS ONTO A WATER SURFACE



TIME, SEC

TABLE I

COMPARISON OF MAXIMUM POOL RADIUS, RMAX, AND TIME TO COMPLETE 'VAPORIZATION, TMAX, AS PREDICTED BY DIFFERENT ANALYSES

LNG SPILL VOLUME-LIQUID

	10 m ³		1000 M ³	
ANALYSIS	RMAX(M)	TMAX(SEC)	RMAX(M)	TMAX(SEC)
LNGVG	20	44	113	111
FAY[5]	16	24	109	108
RAJ[4]	20	38	115	120

REFERENCES

- W. G. May and P. V. K. Perumal, "The Spreading and Evaporation of LNG on Water", ASME Annual Winter Meeting, Nov. 17-22, 1974, N.Y., N.Y.
- 2. Boyle, G. J., and Kneebone, A., "Laboratory Investigations into the Characteristics of LNG Spills on Water: Evaporation, Spreading, and Vapor Dispersion", Shell Research, Ltd., Report to A.P.I. Project on LNG Spills on Water, Ref. 6Z32, March 1973.
- 3. G. E. Feldhauer, et al., "Spills of LNG on Water-Vaporization and Downwind Drift of Combustible Mixtures", Esso Research and Eng., Co., Report No. EEGIE-72, Pg. 52; 24 May 1972.
- 4. P. K. Raj and A. S. Kalelkar, "Fire Hazard Presented by a Spreading, Burning Pool of Liquified Natural Gas on Water', Paper No. 73-25, Page 8, Western States Section/The Combustion Institute 1973 Fall Meeting.
- 5. J. A Fay, "Unusual Fire Hazard of LNG Tanker Spills", Combustion Science and Technology, 1973, Vol. 7, pp. 47-49.

```
PROGRAM LNDVG(LNGIN, TAPE2=LNGIN, LNGGUT, TAPE3=LNGGUT)

CALL DEVICE (GHCREATE, GHLNGGUT, 30000)

A = REGRESSION RATE OF CONSTITUENT 1 OF THE LNG, FT/SEC

AB = USED IN CALCULATING A

CON = CONSTANTIO, 6759

CELOCON = SPENITY OISE, BETWEEN WATER AND LNG ON WATER, LBM/CU.FT.

DELVCI) = SPENITY OISE, BETWEEN WATER AND LNG ON WATER, LBM/CU.FT.

DELVCI) = VOLUME OF CONSTITUENT 1 VAPORIZED DURING A TIME STEP, CU.FT

DENSI = SUMMATION OF DENSITY STORY

DENSI = SUMMATION OF CONSTITUENT IN STORY

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C IN UNITS OF CU.FT.

4 DIMENSION NUMBER(5), FRI(5), RHO(5), HVAP(5), TVAP(5), CP(5), QB(5), Q(5)

1, VOL(5), QBFT(5), DELV(5), FR(5), FRI(5), DLVS(5), TDLV(5)

C READ INPUT DATA
6 READ(2,1000) INST, ICONT, NOSPE, N1, N2, N3, STEP1, STEP2, STEP3, G, RHOW,
1RSPIL, SPILT, VOLI, TNOT, REGR, HBRK, ROAV, CON, PTIM, M2, M3, M4
WRITE(3,1020)
WRITE(3,1020)
WRITE(3,1030)
WRITE(3,1030)
WRITE(3,1040) INST, ICONT, NOSPE, N1, N2, N3, STEP1, STEP2, STEP3, G, RHOW,
1RSPIL, SPILT, VOLI, TNOT, REGR, HBRK, ROAV, PTIM, M2, M3, M4, CON
WRITE(3,1050)
7 DO 20 I=1, NOSPE
8 READ(2,1010) NUMBER(I), FRI(I), RHO(I), HVAP(I), TVAP(I), CP(I)
9 WRITE(3,1060) NUMBER(I), FRI(I), RHO(I), HVAP(I), TVAP(I), CP(I)
20 CONTINUE
AB=0.0
                  20 CONTINUE

AB=0.0

INITIALIZE PARAMETERS

22 DG 30 I=1,NGSPE

23 GBFT(I)=(CP(I)*(TVAP(I)-TNGT)+HVAP(I))*RHG(I)

24 VGL(I)=FRI(I)*VGLI

25 AB=AB+(QBFT(I)/QBFT(I))*VGL(I)/VGLI

26 QB(I)=QBFT(1)*3.14/QBFT(I)

TDLV(I) = 0.000

30 CONTINUE

WRITE(3.1070)
                                    CONTINUE
WRITE(3,1070)
R=(VOL1/3,14)**0.3333
DELRO=RHOW-ROAV
                                    V=VOLT
H=R
A=REGR/AB
IF(ICONT.EQ.1) RMAX=(RSPIL/(3.14*REGR))**0.5
STEP=STEP1
                                    N=N1+N2+N3
                  N=N1+N2+N3

N2=N2+N1

TVOL = 0.0000

PT1 = 0.0000

TIM = 0.0000

PT=PTIM

IDOT = 0

IF(INST.EQ.1) RMAX=9999.

WRITE(3,1080) TIM,N,RMAX,R,H,DELRO,A

35 DO 40 I=1,NOSPE

36 Q(I)=QB(I)*A

WRITE(3,1090) I,VOL(I),QBFT(I),QB(I)

40 CONTINUE

WRITE(3,1100)
              40 CONTINUE
WRITE(3,1100)
TRANSIENT CONTINUOUS OR INSTANTANEOUS SPILL CALCULATIONS
DO 350 [=1, N]
IF(I.GT.N1) STEP=STEP2
IF(I.GT.N2) STEP=STEP3
TIM=TIM+STEP
IF(TIM.GT.SPILT) RSPIL=0.0
IF(R.EQ.RMAX) GO TO 305
303 RDOT=CON*((G*DELRO*/RHOW)**0.25)*(V**0.25) /(TIM**.5)
304 R=R+RDOT*STEP
IF(R.GE.RMAX) R=RMAX
IF(R.GE.RMAX) RDOT = 0.000
IF(R.EQ.RMAX) IDOT = 1
305 TDELV=0.0
   C
               305 TDELV=0.0
```

```
VA=0.0
DENS=0.0
DD 310 J=1,NOSPE
306 DELV(J)=Q(J)*VOL(J)*(R**2)*STEP/V
307 VOL(J)=VOL(J)+RSPIL*FRI(J)*STEP-DELV(J)
308 TDELV=TDELV+DELV(J)
VA=VA+VOL(J)
309 DENS=DENS+RHO(J)*VOL(J)
DLVS(J) = DELV(J)/STEP
310 CONTINUE
TDLVS = TDELV/STEP
TVOL = TVOL + TDELV
V=VA
TVOL = TVOL + TDELV

V=VA

311 DENS1=DENS/V

DELRO=RHOW-DENS1

313 H=V/(3.14*(R**2))

331 DO 340 M=1,NOSPE

332 FRII (M)=VOL(M)/V

333 FR(M)=DELV(M)/TDELV

TDLV(M) = TDLV(M) + DELV(M)

340 CONTINUE

IF(1.GT.N1) PTIM=PT*M2

IF(1.GT.N2) PTIM=PT*M3

PT1=PT1+STEP

IF(1DOT.EQ.1) GO TO 347

IF(H.LE.HBRK) GO TO 347

IF(PT1.GE.PTIM) GO TO 347

GO TO 349

347 CONTINUE

WRITE(3,1110) I,TIM,R,H,DENS
 347 CONTINUE

WRITE(3,1110) I,TIM,R,H,DENS1,RDOT

WRITE(3,1115)

WRITE(3,1120) (J,VOL(J),DLVS(J),TDLV(J),FRII(J),FR(J),J=1,NOSPE)

WRITE(3,1130) V,TDLVS,TVOL

IF(IDOT.EQ.1) GO TO 348

PTI = 0.0000

348 IDOT = 0

349 CONTINUE

IF(H.LE.HBRK) GO TO 600

350 CONTINUE

OR RMAX=R
IF(H.LE.HBRK) GD 10 600
350 CONTINUE
600 RMAX=R
WRITE(3,1200)
WRITE(3,1210) RMAX,H,TIM
PTIM=PT*M4
K=1
602 DO 650 1=K,N
IF(I.GT.N1) STEP=STEP2
IF(I.GT.N2) STEP=STEP3
TIM=TIM+STEP
604 RBRK=(RMAX**2-(V/(HBRK*3.14)))**0.5
RMK=(RMAX-RBRK)/RMAX
IF(RMK.LT.0.01) GO TO 1660
VA=0.0
TDELV=0.0
606 DO 610 J=1,NOSPE
607 DELV(J)=Q(J)*(VOL(J)/V)*(RMAX**2-RBRK**2)*STEP
VOL(J)=VOL(J)-DELV(J)
IF(VOL(J).LT.0.0) VOL(J)=0.0
VA=VA+VOL(J)
TDELV=TDELV+DELV(J)
DLVS(J) = DELV(J)/STEP
```

* . 1

```
610 CGNTINUE
TOLYS = TOELV/STEP
TVOL = TVOL + TDELV
TOLV(M) = TOLV(M) / TDEV
612 FR(M)-DELV(M)/TDEV
613 FRII(M)=VOL(M)/V
TOLV(M) = TDLV(M) + DELV(M)
620 CONTINUE
620 CONTINUE
630 CONTINUE
630 CONTINUE
640 CONTINUE
650 CONTI
```

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